Magnetization of ultrathin Fe films deposited on Gd (0001)

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(Received 29 September 1997; accepted 26 October 1997)

The magnetization of ultrathin (1.5–6 ML) Fe films deposited on Gd (0001) was studied using spin polarized secondary electron emission spectroscopy. An anomalously high Curie temperature (>320 K) was observed for very low coverages of Fe (1.5 ML). The anomalously high Curie temperature results in part from the strong antiferromagnetic exchange interaction between the Fe and Gd at the interface. In addition, a spin reorientation transition of the Fe magnetization from in-plane to perpendicular was observed as the temperature is increased for Fe thickness between 1.5–4 ML. The spin reorientation is attributed to the competition between the perpendicular anisotropy of the Fe overlayer and the exchange interaction with the in-plane Gd bulk magnetization. Therefore the effects of varying the thickness of the Gd and hence the bulk Curie temperature on the spin reorientation transition can be studied. [S0734-2101(98)00303-0]

The rare-earth transition metal (RE-TM) compounds are of significant scientific and technological importance due to their versatile magnetic properties and strong perpendicular anisotropy.[1] The magnetic properties of these compounds are determined by the strong antiferromagnetic coupling between the $4f$ magnetic moments of the RE and the $3d$ magnetic moment of the TM. This antiferromagnetic coupling is mediated by the overlap of the $5d$ conduction electrons of the RE with the $3d$ magnetic moment of the TM. In addition, the antiferromagnetic coupling of the RE-TM pairs and the ferromagnetic coupling of the RE-RE and the TM-TM pairs results in a ferrimagnetic system. Finally, these compounds exhibit a strong anisotropy perpendicular to the surface. The origin of this anisotropy is not fully understood;[2] however, recent structural studies suggests that higher perpendicular versus in-plane correlations of the RE-TM pairs is related to the magnetic anisotropy.[3]

In an attempt to understand the nature of the RE-TM antiferromagnetic coupling the magnetization of ultrathin Gd films deposited on the Fe (100) surface was studied using spin polarized Auger electron spectroscopy (SPAES).[4] In this study the Curie temperature of the magnetization at the Gd/Fe interface was found to be ~ 800 K; however, this large Curie temperature is driven by the Curie temperature of the underlying Fe substrate, and is too high to have any practical applications in magneto-optic storage devices. A more interesting application would be to deposite ultrathin Fe films on Gd. This would allow the relatively low Curie temperature of the Gd ($T_C = 293$ K) to be exploited.

In this study ultrathin films (1.5–6 ML) of Fe are deposited on a Gd (0001) base layer at low temperature. The Gd (0001) base layer was grown on a single crystal Y (0001) substrate, and the necessary growth conditions for a smooth, well ordered Gd surface were established. A complete loss of the low-energy electron diffraction LEED pattern is observed at very low coverages of Fe, evidence that a disordered Fe overlayer is formed. The magnetization of the Fe/Gd system was monitored *in situ* using spin polarized secondary electron emission spectroscopy (SPSEES). An anomalously high Curie temperature was observed for very low coverages of Fe (1.5 ML). In addition, a spin reorientation of the Fe magnetization was observed slightly below the Gd bulk Curie temperature, with an accompanying loss of long range magnetic order in the Gd bulk.

The experiment was conducted in an ultrahigh vacuum chamber with a base pressure of 5×10^{-11} Torr. The SPSEES is performed by bombarding the sample with unpolarized 1 keV primary electrons. The primary electrons eject secondary electrons from the sample, that are then spin analyzed, giving a signal proportional to the average magnetic moment of the first few atomic layers of the surface. The polarization is measured as follows: the sample was magnetized by the magnetic field (~ 250 G) generated by a single current pulse passed through a coil with its axis oriented parallel to the substrate face (x axis). The magnetic field was first applied in the positive x direction ($\uparrow$) and the corresponding left–right intensities ($I_L^{\uparrow}$ and $I_R^{\uparrow}$) were measured by the electron spin analyzer. The magnetic field was then applied in the negative x direction ($\downarrow$) (reverse the current pulse) and the corresponding left-right intensities ($I_L^{\downarrow}$ and $I_R^{\downarrow}$) were again measured by the electron spin analyzer. The polarization, P, was then calculated using

$$P = \frac{1}{S_{\text{eff}}}\frac{A-1}{A+1},$$

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where A is the Mott scattering asymmetry[5] given by

$$A = \sqrt{\frac{I_R^{\uparrow} I_L^{\downarrow}}{I_L^{\uparrow} I_R^{\downarrow}}},$$

and S_{eff} is the effective Sherman function. This procedure is used to eliminate any instrumental asymmetries present in the electron spin analyzer. All measurements were performed in remanance, i.e, in the absence of an applied magnetic field. The polarization was then measured as a function of temperature. The sample was cooled by a liquid N_2 reservoir and was heated radiatively by a tungsten filament.

Yttrium was chosen as a substrate for growing smooth Gd films rather than tungsten because the Gd (0001)/Y (0001) lattice mismatch ($<0.5\%$) is much smaller than the Gd (0001)/W (110) lattice mismatch ($>12.9\%$). In addition, the surface free energy for yttrium is only slightly larger than that of gadolinium whereas the surface free energy for tungsten is four times larger.[6] Therefore the complications brought on by strain in the Gd/W system can be circumvented by growing Gd on Y (0001). Given the low base pressure of the chamber the yttrium surface is no more difficult to clean that of the tungsten. To remove trace contaminants in the surface region, the yttrium substrate was repeatedly sputtered with argon at 2 keV (8×10^{-6}A) and subsequent anneals to 973 K, for 10 min. In order to preserve the smoothness of the Y surface, the substrate was rotated $\pm15°$ after each sputter/anneal cycle to a maximum angle of $\pm45°$ away from normal incidence. This procedure was repeated until the only remaining contamination was carbon and oxygen. The sputter/anneal cycles were then reduced to sputtering at 0.5 keV (2×10^{-6}A), and subsequent flash annealing to 973 K. This resulted in the sharp (1×1) sixfold LEED pattern expected for the hcp face, with the only trace of contamination being oxygen at the 10% level as determined by AES.

The Gd was deposited onto the clean Y (0001) substrate by sublimation from 99.99% pure Gd lump, packed into a tungsten crucible, which was e-beam heated in a water cooled cell. During deposition, the substrate was held at 300 K to promote layer-by-layer growth of the Gd. The Gd thickness was determined using a quartz crystal microbalance. Gd thicknesses of 115 Å ($\sim$40 ML) were primarily used for this study so that the bulk Curie temperature, T_{CB}, of 293 K is realized[7] and the shape anisotropy is sufficient to hold the magnetization in-plane.[8] The as-deposited Gd film was then flash annealed to 723 K, giving the sharp (1×1), sixfold LEED pattern expected for the hcp face with no trace of contamination. In Fig. 1(a), the secondary electron spin polarization versus temperature for a smooth, well ordered Gd surface is shown. As can be seen, the Curie temperature of the Gd film is 291 ± 1 K. No evidence for an enhanced surface Curie temperature was found for the Gd surface. One possible reason for this could be the absence of the strain induced anisotropy at the Gd surface, which results from the large lattice mismatch at the Gd/W interface.[9] Moreover, the magnetic properties of the Gd (0001)/Y (0001) system are much more reproducible than those of the Gd (0001)/W (110) system. This can be attributed to the lack of strain in the Gd/Y system.

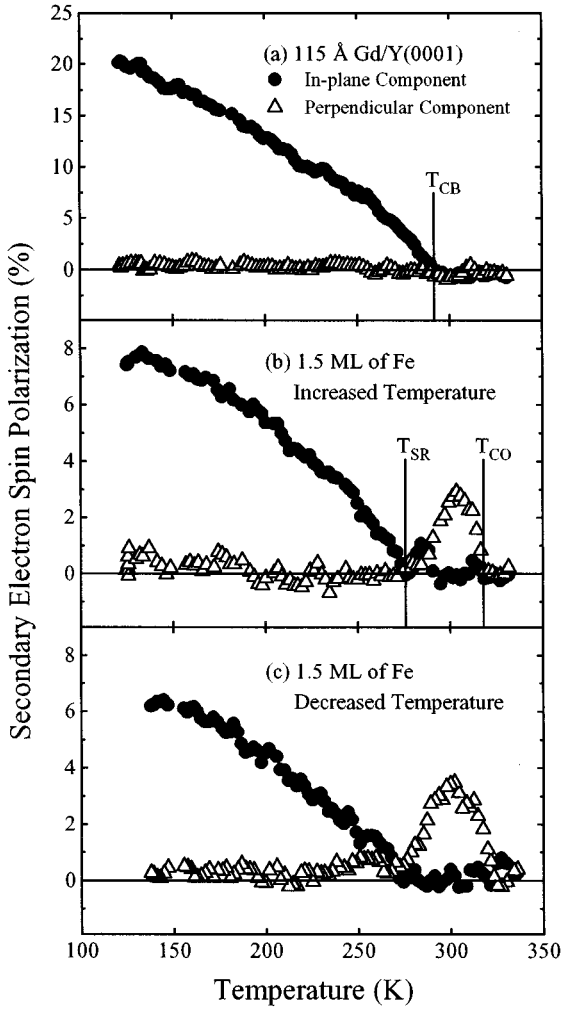

FIG. 1. (a) Secondary electron spin polarization vs temperature for a 115 Å Gd film grown on Y (0001). (b) and (c) the spin reorientation transition for a 1.5 ± 0.5 ML Fe film deposited on the Gd (0001) surface. In panel (b) the temperature is ramped up and in panel (c) the temperature is ramped down demonstrating the reversibility of the transition. The sample was remanently magnetized at each data point.

The Fe was then deposited onto the Gd (0001) surface by sublimation from a 99.999% pure Fe ingot which was e-beam heated in a water cooled cell. During deposition the Gd base layer was held at 100 K to prevent intermixing between the Fe and Gd. The Fe thickness was determined using a quartz crystal microbalance. Although expitaxial growth of Fe on Gd (0001) is not possible due to geometry constraints and the large lattice mismatch, the Fe overlayer does grow homogeneously. This is evident by an increase in the Fe Auger signal and a proportional decrease in the Gd Auger signal with increasing Fe coverage. In addition, the Fe/Gd (0001) LEED pattern is completely quenched at very low coverages of Fe (<1.5 Å $=1$ ML), indicating that island formation or alloying does not occur.

In Fig. 1(b) the secondary electron spin polarization ver-

sus temperature for a 1.5 ML Fe overlayer on Gd (0001) is shown. The overall reduction of the in-plane polarization below the Gd bulk Curie temperature demonstrates that the magnetic moments of the Fe atoms are oriented antiparallel to the Gd bulk magnetization, as expected for the RE-TM antiferromagnetic coupling. As can be seen, the polarization switches from in-plane to perpendicular as the temperature is increased. The transition occurs slightly below the Gd bulk Curie temperature, demonstrating that a spin reorientation transition, T_{SR}, of the Fe overlayer takes place at ~ 275 K. This spin reorientation transition results from the competition of the perpendicular surface anisotropy of the Fe overlayer and the exchange interaction of the overlayer with the Gd bulk magnetization and is inherently different from the spin reorientation transition observed for the $3d$ transition metals which results from a temperature dependent anisotropy.[10] A 3.5% perpendicular component of the in-plane polarization is observed up to the overlayer Curie temperature, T_{CO}, of 320 K. This Curie temperature is much higher than the Curie temperature expected for a 1.5 ML Fe film [$T_C \approx 150$ K (Ref. 11)], demonstrating that the interfacial Gd atoms are participating in the magnetic ordering above T_{CB}. In panel (c) the spin reorientation transition is shown to be completely reversible upon decreasing the temperature.

A more complete study of the temperature and thickness dependence of the magnetization of ultrathin Fe overlayers deposited on the Gd (0001) surface is shown in Fig. 2, where the magnetization for successive thickness of Fe is plotted. In panel (b), the secondary electron spin polarization versus temperature for a 2 ML Fe overlayer is shown. Again the in-plane polarization is reduced at low temperatures due to the antiferromagnetic coupling of the Fe and Gd. The in-plane polarization at low temperature continues to decrease with increasing Fe thickness and crosses zero between 2 and 4 ML, as shown in panel (c). The change in sign of the in-plane polarization observed from panel (b) to panel (c) can be attributed to the sensitivity of the SPSEES to the Fe magnetization. The spin reorientation transition is evident for the 2 and 4 ML thick Fe overlayers, as shown in panels (b) and (c). An important aspect of this data is that the spin reorientation transition occurs at temperatures less than the Gd bulk Curie temperature, i.e., $T_{SR} < T_{CB}$. This is because (1) the Fe–Gd exchange interaction is greater than the Gd–Gd exchange (-2.2×10^{-15} erg vs 0.5×10^{-15} erg),[2] and (2) the Fe overlayer is coupled to the Gd bulk magnetization via the Fe–Gd exchange interaction. Because the Gd bulk magnetization is held in-plane by its shape anisotropy, a 90° domain wall can exist between the overlayer and Gd bulk magnetization, for $T_{SR} < T < T_{CB}$. From an analysis of the domain wall energy[12] using the Gd–Gd exchange energy of 0.5×10^{-15} erg and a Gd bulk anisotropy of $K_{lu} \approx 10^5$ erg/cm^3, an estimate of the domain wall width of ~ 40 Å is obtained. This width is less than the 115 Å Gd film thickness used in this study. The presence of a domain wall would explain the reduction of the total polarization observed between T_{SR} and T_{CB} in panels (b) and (c). For a 6 ML thick Fe overlayer, Fig. 2(d), the dipole energy of the Fe is strong enough to hold the magnetization in the plane of the film. The change in sign of the in-plane magnetization at T_{CB} is due to the fact that the film is magnetized at every temperature. Thus, when the Gd base layer goes paramagnetic, the magnetic moments of the Fe overlayer will align with the applied magnetic field, resulting in a positive signal. The important consequence of the in-plane remanance for the 6 ML films is that it is nearly the same as the critical thickness

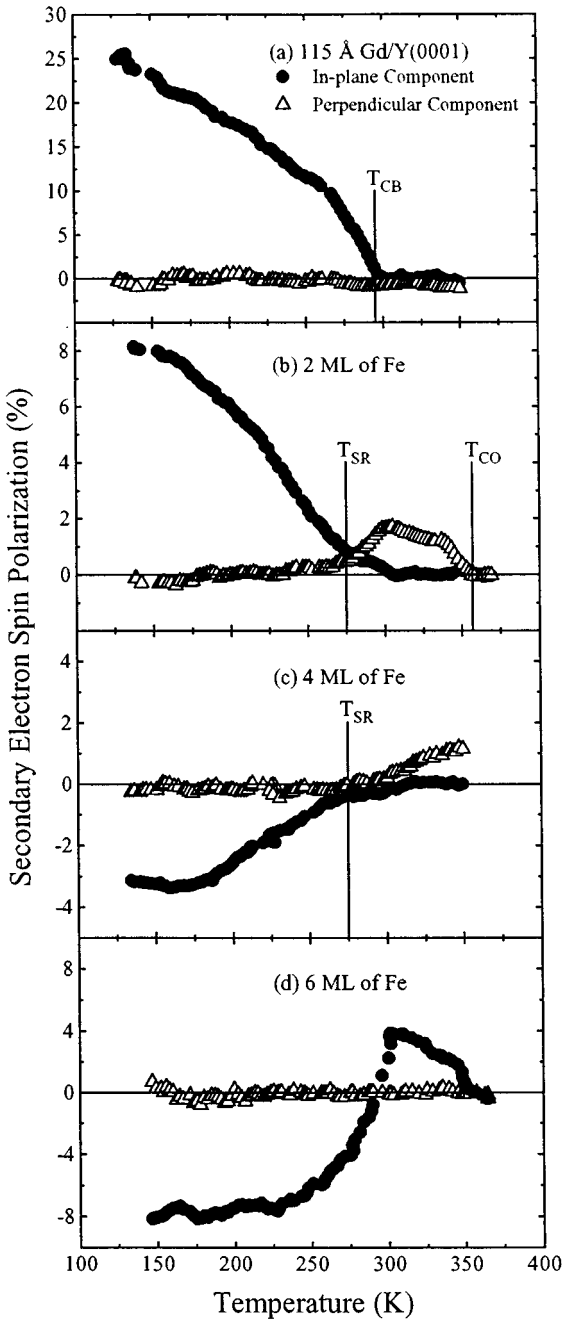

FIG. 2. (a) Secondary electron spin polarization vs temperature for increasing thickness of Fe deposited on the Gd (0001) surface. The data is completely reversible on decreasing the temperature. The sample was remanently magnetized at each data point.

for single crystal bcc- and fcc Fe films grown on Ag (001) and Cu (001) substrates. This shows that the strength of the uniaxial perpendicular anisotropy is very close to that of the single crystal films, i.e., ~ 0.5 erg/cm^2 (Ref. 11) if the magnetic moments of the Fe atoms in these disordered films are similar to that of the single crystal films. Figures 2(a)–2(d) therefore demonstrate the antiferromagnetic nature of the Fe–Gd exchange interaction, the surface sensitivity of the secondary electron spin polarization, the magnitude of the uniaxial anisotropy in the Fe films, and the Fe spin reorientation transition.

In conclusion, the magnetization of ultrathin (1.5–6 ML) Fe films deposited on the Gd (0001) surface was studied. An anomalously high Curie temperature ($T_C > 320$ K) was observed for very low coverages of Fe (1.5 ML) indicating that the interfacial Gd atoms are participating in the magnetic order of the Fe above the Gd bulk Curie temperature. In addition, a reversible spin reorientation transition from in-plane to perpendicular magnetization was observed as the temperature is increased for Fe thickness between 1.5–4 ML. The onset of this transition is observed well below the Gd bulk Curie temperature and appears to be accompanied by the formation of a domain wall in the vicinity of T_{SR}. The spin reorientation transition is attributed to the competition of the perpendicular surface anisotropy of the Fe overlayer and the exchange interaction of the overlayer with the in-plane Gd bulk magnetization. This study opens an avenue of research for designing the structural and magnetic properties of the important RE-TM compounds.

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